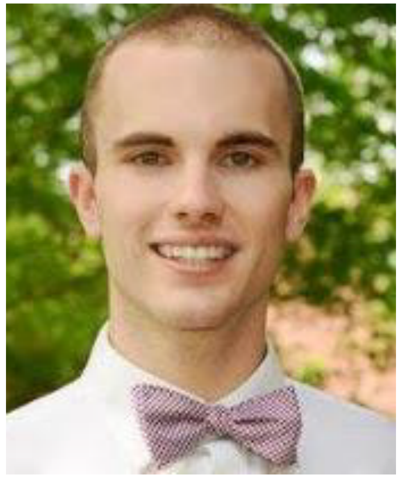
# Chiral magnetism in oxide interfaces & 2D materials

Mohit Randeria
The Ohio State University

Correlated Oxides
& Oxide Interfaces,
Minnesota, May 2014

MRSEC

Sumilan Banerjee



James Rowland

Onur Erten
(now at Rutgers)

MRSEC

* Ferromagnetic exchange, spin–orbit coupling and spiral magnetism at the $LaAlO_3/SrTiO_3$ interface
  S. Banerjee, O. Erten & MR, Nature Physics **9**, 626 (2013)

* Skyrmions in 2D chiral magnets
  S. Banerjee, J. Rowland, O. Erten & MR, arXiv: 1402.7082

# Outline:

- ❑ Oxide interfaces: evidence for magnetism
  $LaAlO_3/SrTiO_3$ (LAO/STO)

- ❑ Broken inversion + Spin-orbit coupling:
  Symmetry → chiral magnetic interactions
  & compass anisotropy

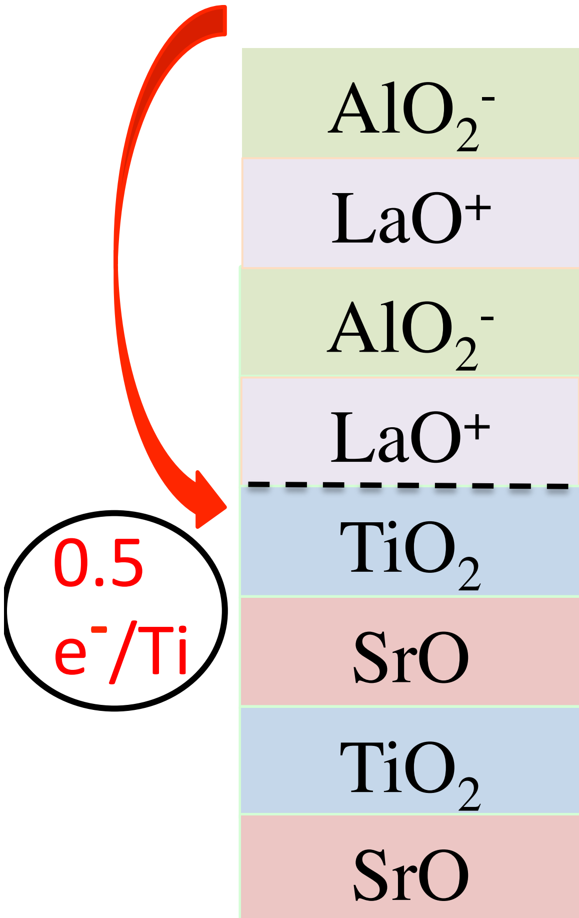
- ❑ Microscopic considerations:
  exchange, DM & anisotropy

- ❑ Spiral ground state in H=0

- ❑ Skyrmion crystal in finite field
  in 2D chiral magnets

# LAO/STO Interface

## Polar catastrophe




$LaAlO_3$

$E_g$ = 5.6 eV

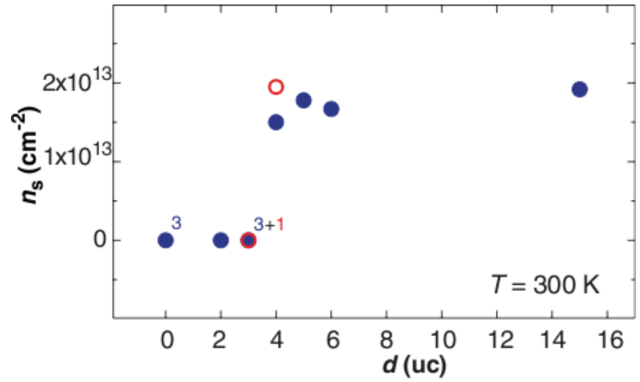
n-type
interface

$SrTiO_3$

$E_g$ = 3.2 eV

Ohtomo & Hwang,
Nature (2004)

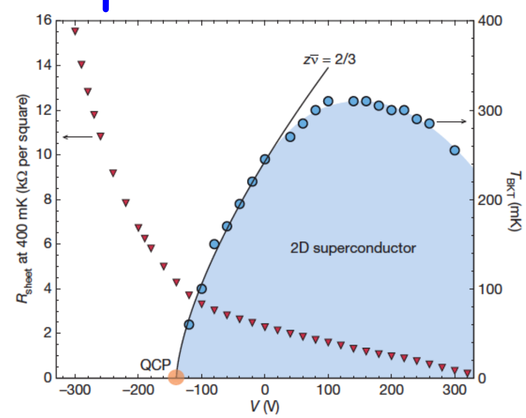
## 2DEG




Thiel et al,
Science
(2006)

Hall → n

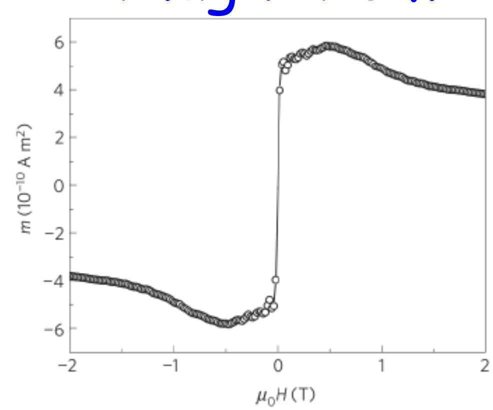
## Superconductivity




Caviglia et al,
Nature
(2008)

$T_c(n)$

## Magnetism

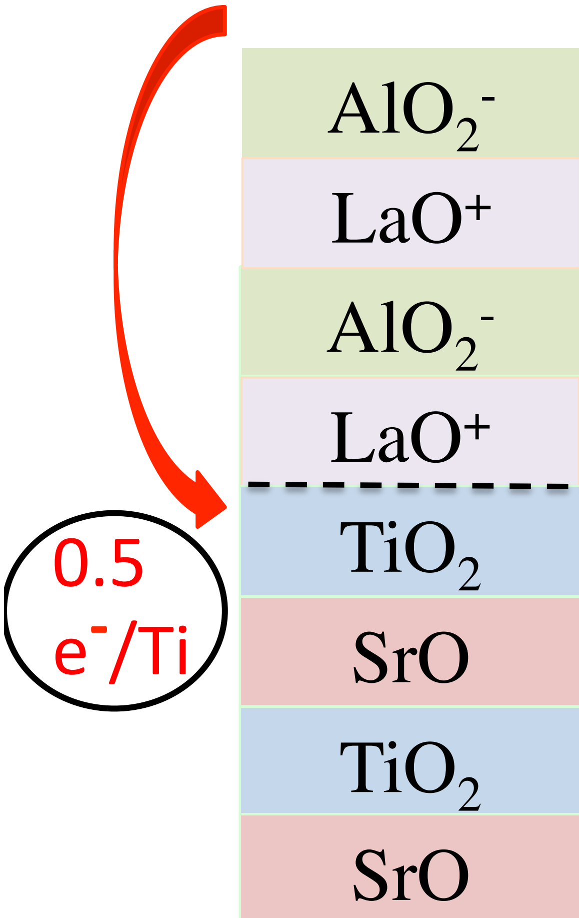



Li et al,
Nat. Phys
(2008)

M(H)

# LAO/STO Interface

Polar catastrophe




$LaAlO_3$

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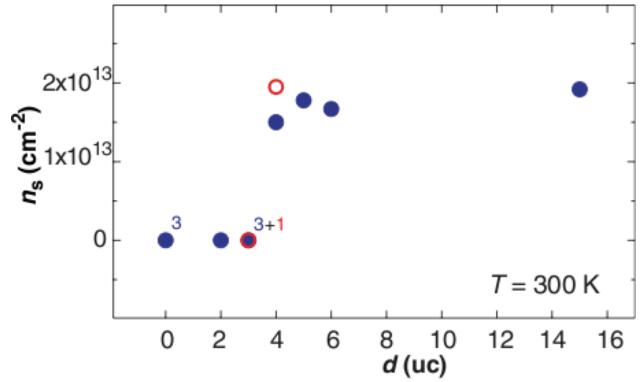
Ohtomo & Hwang, Nature (2004)

## 2DEG




Thiel et al, Science (2006)

Hall → n

Polar Catastrophe

$= n_{2D} \sim 3.3 \times 10^{14}$ cm$^{-2}$

+ additional electrons from O-vacancies

Hall → mobile carriers

$n_{2D} = 2\text{x}10^{13}$ cm$^{-2}$

~ 10% of polar catastrophe

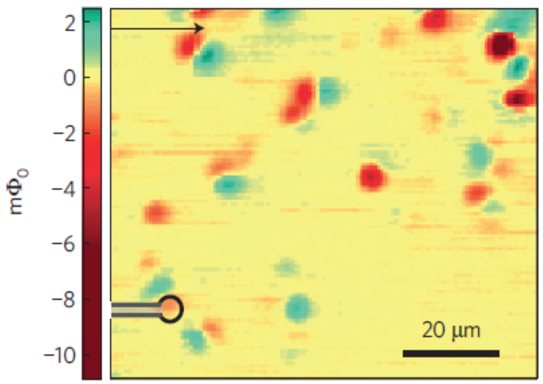
*cf.* 100% in GTO/STO [Stemmer]

Where are the missing electrons in LAO/STO?

# If the polar catastrophe occurs in LAO/STO, where are ~ 90% electrons not seen in transport?

Answers -- not certain
-- details are sample & probe dependent

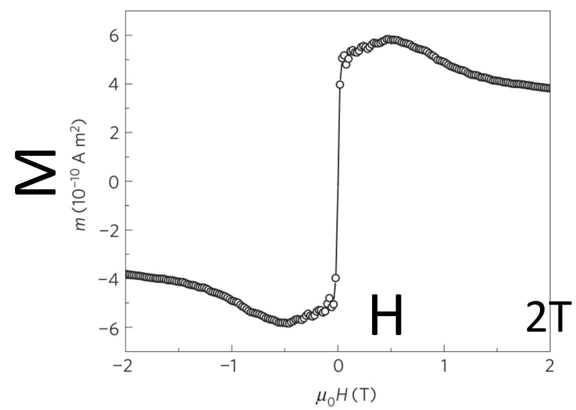
Many experiments → large density of Localized electrons that behave like local moments




**Scanning SQUID**

Bert et al. Nature Phys. (2011)

* Inhomogeneous; M=0
* Isolated micron-size patches of in-plane FM
* Susceptometry: local moments ~ 0.5 e/Ti




**Torque Magnetometry**

Li et al, Nature Phys (2011)

* M ≈ 0.3 - 0.4$\mu_B$ /Ti
* Exchange scale ~ 100K
* No hysteresis

**Spectroscopy: Ti $d^1$ states**

XPS Sing et al, PRL (2009)

XMCD ~ 0.1 e/Ti
Lee et al, Nature Mat. (2013)

# Lessons from LAO/STO experiments

despite sample-dependence & many open questions!

* 2D interface

* Broken inversion  z → -z

* Rashba spin-orbit coupling (SOC)

$$\mathcal{H}_{\rm soc} = \lambda_{\rm soc}\, \widehat{\mathbf{z}} \cdot (\mathbf{k} \times \sigma)$$

tunable by gate voltage [Expt: Caviglia et al., PRL (2010)]

* Many experiments see evidence for magnetism

* Disorder & inhomogeneity

which I will ignore

Theory must understand the clean problem first

# Summary of Main Results

* If there is magnetic order at oxide interfaces,
  broken inversion & SOC → chiral magnetism
  independent of
  microscopic details

* long wavelength spiral ground state at H=0
  with pitch ~ gate tunable SOC

* skyrmions in finite field down to T=0

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  in chiral magnets

## Symmetry → Ginzburg-Landau (GL) free energy for local magnetization **m(r)**

Inputs: 2D; broken inversion; SOC

$$\boxed{\begin{aligned}\mathcal{F} = \mathcal{F}_0(\mathbf{m}) &+ (J/2)\sum_\alpha (\nabla m^\alpha)^2 \\ &-D[(m^z\partial_x m^x - m^x\partial_x m^z) - (m^y\partial_y m^z - m^z\partial_y m^y)] \\ &-A_c[(m^x)^2 + (m^y)^2] + \frac{A_c}{2}[(\partial_x m^y)^2 + (\partial_y m^x)^2] \\ &+A_s(m^z)^2\end{aligned}}$$

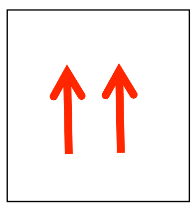
Simpler to explain various terms using "effective spin Hamiltonian"

$$\mathcal{F} = \boxed{\mathcal{F}_0(\mathbf{m}) + (J/2)\sum_{\alpha}(\nabla m^{\alpha})^2}$$
$$-D[(m^z\partial_x m^x - m^x\partial_x m^z) - (m^y\partial_y m^z - m^z\partial_y m^y)]$$
$$-A_c[(m^x)^2 + (m^y)^2] + \frac{A_c}{2}[(\partial_x m^y)^2 + (\partial_y m^x)^2]$$
$$+A_s(m^z)^2$$

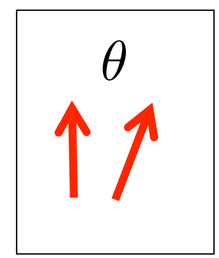
$$\boxed{\mathcal{H}_{\mathrm{J}} = -J\sum_{(i,j)}\mathbf{S}_i\cdot\mathbf{S}_j}$$

Isotropic exchange J

$J > 0 \Rightarrow$ Ferromagnet



$$E_J(\theta) = -J\mathbf{S}_i\cdot\mathbf{S}_j$$
$$\sim -J\cos\theta \sim J\theta^2$$




Various microscopic mechanisms:
e.g., Zener double-exchange; RKKY; Stoner

$$\mathcal{F} = \mathcal{F}_0(\mathbf{m}) + (J/2)\sum_\alpha (\nabla m^\alpha)^2$$
$$\boxed{-D[(m^z\partial_x m^x - m^x\partial_x m^z) - (m^y\partial_y m^z - m^z\partial_y m^y)]}$$
$$-A_c[(m^x)^2 + (m^y)^2] + \frac{A_c}{2}[(\partial_x m^y)^2 + (\partial_y m^x)^2]$$
$$+A_s(m^z)^2$$

Chiral exchange: DM term
[Dzyaloshinskii-Moriya]

$$\boxed{\mathcal{H}_{\mathrm{DM}} = \sum_{(i,j)} \mathbf{D}_{ij}\cdot(\mathbf{S}_i \times \mathbf{S}_j)}$$

* Broken inversion → $D_{ij}$
* SOC → coupling of spins to real-space

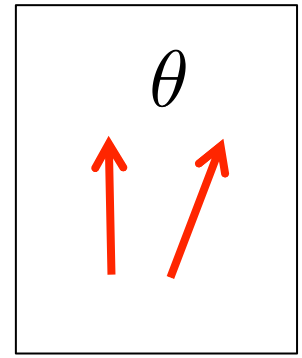
Symmetry → direction of D

$$\boxed{\Rightarrow \mathbf{D}_{ij} \sim \widehat{\mathbf{z}} \times (\mathbf{r}_i - \mathbf{r}_j)}$$

Microscopics → magnitude of D

$$\boxed{\Rightarrow |\mathbf{D}| \sim \lambda_{\mathrm{SOC}} \ll J}$$

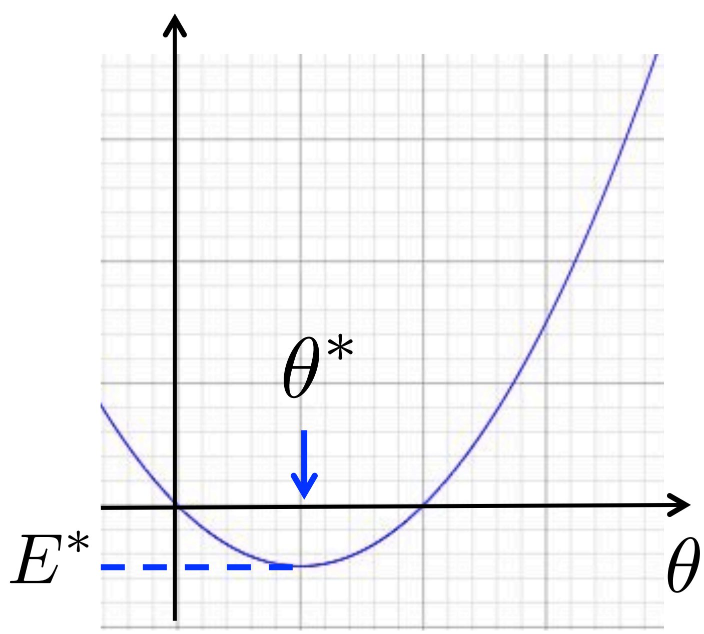
# Ferromagnetic exchange + chiral DM term




$$J\mathbf{S}_i \cdot \mathbf{S}_j \simeq J\theta^2/2$$

$$\mathbf{D}\cdot(\mathbf{S}_i \times \mathbf{S}_j) \simeq D\sin\theta \simeq D\theta$$

$$\boxed{E(\theta) = J\theta^2/2 - D\theta}$$




$$\boxed{\begin{aligned}\theta^* &= D/J \\ E^* &= -D^2/2J\end{aligned}}$$

→ Spin-texture (Spiral or skyrmion) with length scale

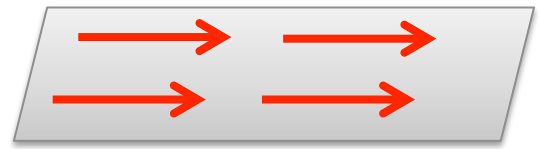
$$\left(\frac{J}{D}\right) a \gg a$$

# Magnetic anisotropy: "Compass" Term $A_c$

$$\boxed{-A_c \sum_i \left(S_i^y S_{i+\hat{x}}^y + S_i^x S_{i+\hat{y}}^x\right)}$$

Broken z-inversion & SOC

* Symmetry allows Kitaev-like term
* Easy-plane anisotropy $\mathbb{Z}_4$



Microscopic origin: <u>same</u> SOC as the DM term

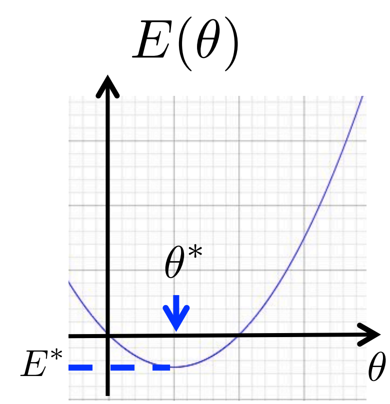
Usually ignored in the literature

$$\boxed{A_c \sim \mathcal{O}\left(\lambda_{\text{soc}}^2\right) \ll D \sim \mathcal{O}\left(\lambda_{\text{soc}}\right)}$$

However, both $A_c$ and D impact energy at the same order.

Cannot ignore $A_c$!

$$\boxed{A_c \sim D^2/J}$$



$$\mathcal{F} = \mathcal{F}_0(\mathbf{m}) + (J/2)\sum_\alpha (\nabla m^\alpha)^2$$ ← Exchange

$$-D[(m^z\partial_x m^x - m^x\partial_x m^z) - (m^y\partial_y m^z - m^z\partial_y m^y)]$$ ← DM

$$\boxed{\begin{aligned}&-A_c[(m^x)^2 + (m^y)^2] + \frac{A_c}{2}[(\partial_x m^y)^2 + (\partial_y m^x)^2]\\ &+A_s(m^z)^2\end{aligned}}$$ ← Anisotropy

Effective magnetic anisotropy: $A = A_c + A_s$

Anisotropy energy ~ $A(m_z)^2$

Compass
$A_c > 0$

single-ion or shape anisotropy -- order of magnitude smaller than compass term in oxide interfaces

A > 0 Easy-plane

A < 0 Easy-axis

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  in 2D chiral magnets

# Microscopic Interlude:

Exchange (J) + DM term (D) + compass anisotropy ($A_c$)

For a wide variety of exchange mechanisms
in both metals & insulators with Rashba SOC

$$\boxed{A_c|J|/D^2 = 1/2} \qquad (\lambda_{\mathrm{soc}} \ll t)$$

* AFM Superexchange + SOC — Moriya, Phys Rev (1960); Shekhtman et al, PRL (1992)
* RKKY + SOC — Imamura et al, PRB (2004)
* Double exchange + SOC — Banerjee, Erten & MR, Nature Phys. (2013)

"Unified" derivation for pair-wise exchange mechanisms

Banerjee, Rowland Erten & MR, arXiv: 1402.7082

# Microscopic Interlude: Double exchange + Rashba SOC

$$\mathcal{H}_0 = -t \sum_{<ij>,\alpha} c^\dagger_{i\alpha} c_{j\alpha} \qquad \mathcal{H}_{\rm int} = -\frac{J_{\rm H}}{2} \sum_{i\alpha\beta} c^\dagger_{i\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{i\beta} \cdot \mathbf{S_i}$$

$$-i\lambda \sum_{<ij>,\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} . \hat{\mathbf{d}}_{ij} c^\dagger_{i\alpha} c_{j\beta} + \text{h.c.} \qquad \hat{\mathbf{d}}_{ij} = \hat{\mathbf{z}} \times \hat{\mathbf{r}}_{ij}$$

## Two-site problem:

$$\mathcal{H}_0 = -\tilde{t} \sum_{\alpha\beta} (c^\dagger_{i\alpha} [e^{i\vartheta \boldsymbol{\sigma} . \hat{d}_{ij}}]_{\alpha\beta} c_{j\beta} + \text{h.c.}) \qquad \tilde{t} = \sqrt{t^2 + \lambda^2} \quad \tan\vartheta = \lambda / t$$

SU(2) "Gauge Transformation" $\quad a_{i\alpha} = [e^{-i(\vartheta/2)\boldsymbol{\sigma}.\hat{d}_{ij}}]_{\alpha\beta} c_{i\beta}$

Anderson-Hasegawa with SOC

$$\mathcal{H}_{\rm DE} = -J_{\rm F} \sum_{<ij>} \left[ 1 + \mathbf{S_i} \cdot \mathcal{R}(\mathbf{2}\vartheta \hat{\mathbf{d}}_{\mathbf{ij}}) \mathbf{S_j} \right]^{1/2}$$

$$\Rightarrow J = J_{\rm F} \cos 2\vartheta \qquad D = J_{\rm F} \sin 2\vartheta \qquad A_c = J_{\rm F}(1 - \cos 2\vartheta)$$

$$\boxed{A_c J / D^2 = 1/2 \quad \text{for} \quad \lambda \ll t}$$

# Summary of Microscopic Interlude:

## Hierarchy of energy scales

| Isotropic FM Exchange | $\gg$ | Chiral DM Exchange | $\gg$ | Compass Anisotropy |
|---|---|---|---|---|

$$J \gg D \gg A_c$$

$$D \downarrow \mathcal{O}(\lambda_{\rm soc}) \qquad A_c \downarrow \mathcal{O}(\lambda_{\rm soc}^2)$$

$$\boxed{A_c J/D^2 = 1/2}$$

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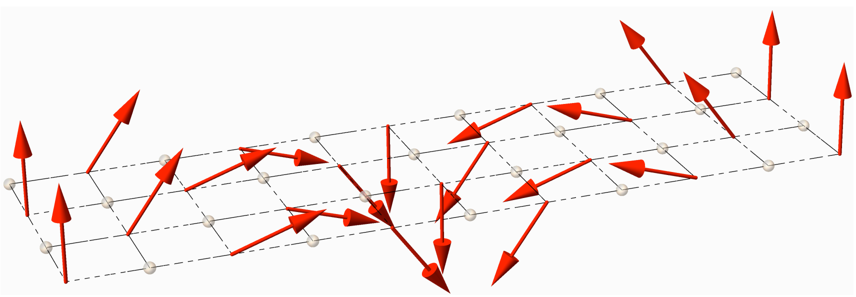
# Solution of GL theory: variational & Monte Carlo

H=0 ground state:

Spiral $AJ/D^2 \lesssim 1$



Spiral pitch is gate-tunable

$$\frac{2\pi}{Q_0} \sim \frac{J}{D} \sim \frac{t}{\lambda_{\mathrm{soc}}}$$

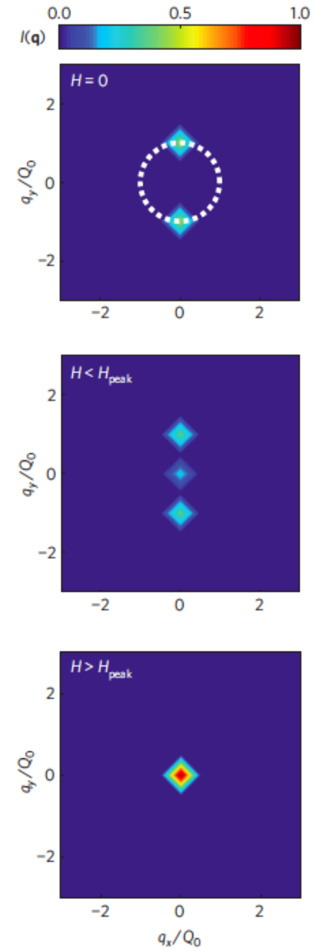
Typical wavelength $2\pi/Q_0 \sim 600\mathring{A}$

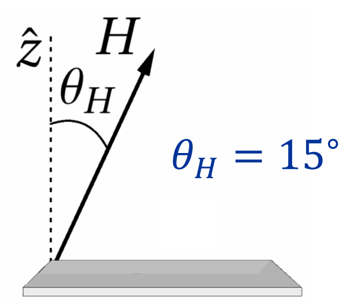
Spin structure Factor I(**Q**)

spiral

FM

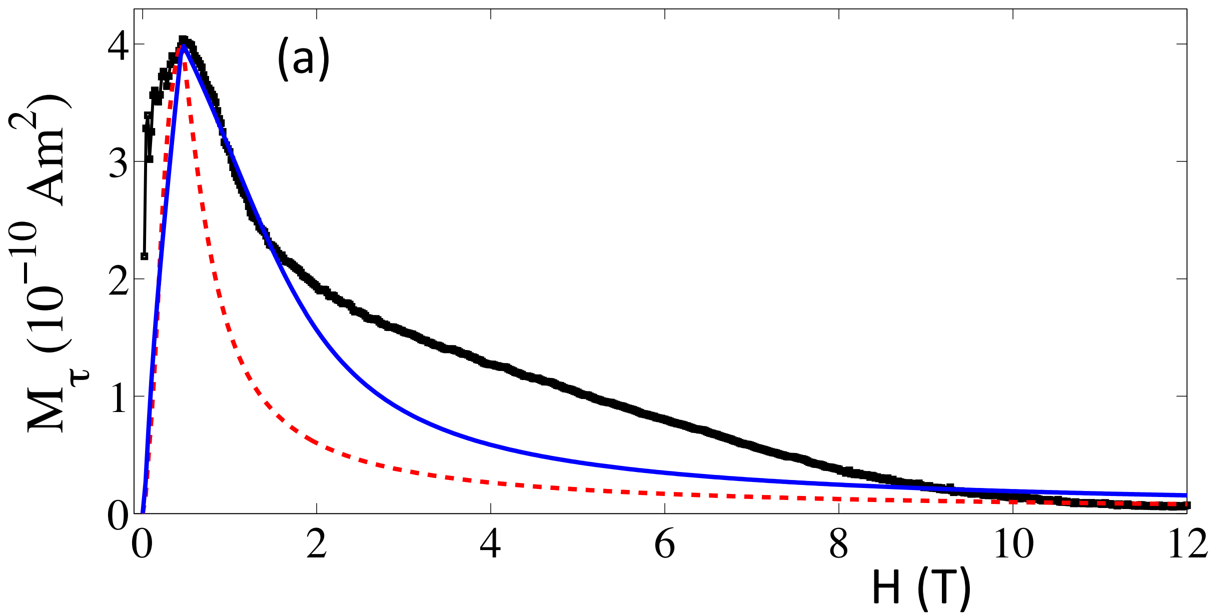
increasing H




Note: L. Li's Expt. Torque geometry

# Comparison with Torque Magnetometry




**Torque:**

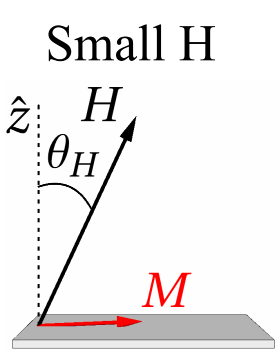
$$\vec{\tau} = \vec{M} \times \vec{H}$$

**Magnetization**

$$M_\tau = \tau / H$$

Spiral with

$M \neq 0$




Expt* Li et al., Nat. Phys. (2011)

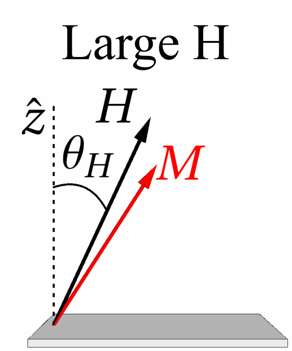
Red: Microscopic fit $AJ/D^2 = 1/2$

Blue: Phenomenological fit $AJ/D^2 = 0.8$

*after background subtraction [with Lu Li]

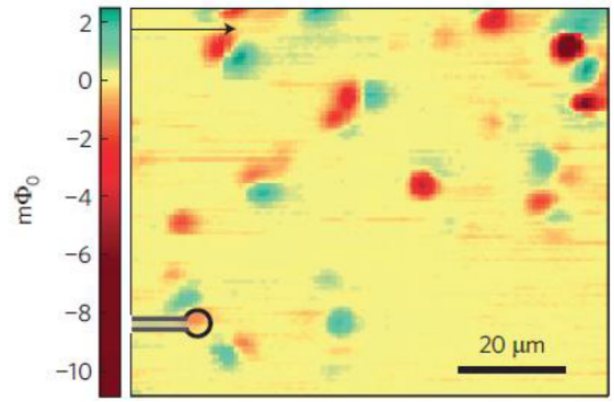
FM with

$M \parallel H$



- Torque magnetometry ✓
- Scanning SQUID

coplanar spiral state has zero net moment at H=0




Bert *et al.* Nature Phys. (2011)

→ Inhomogeneity?

Disorder effects beyond present theory

→ FM patches? [magnetoelastic coupling – see our Nature Phys. (2013)]

→ random in-plane orientation ? Compass anisotropy

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Microscopic Model for LAO/STO: not discussed in this talk!

see: Nature Phys. **9**, 626 (2013)

- Local moment formation: charge localization in top layer $d_{xy}$
- Double exchange: $d_{xy}$ moments + conduction electrons in quasi-1D $d_{xz}$, $d_{yz}$ bands with Rashba SOC

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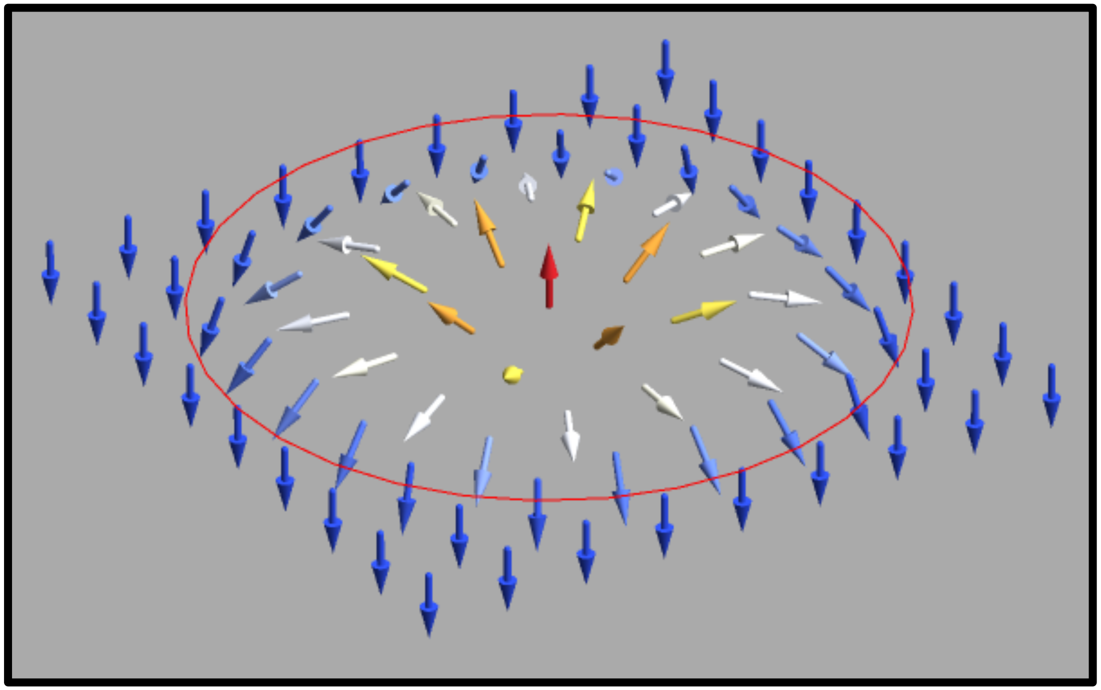
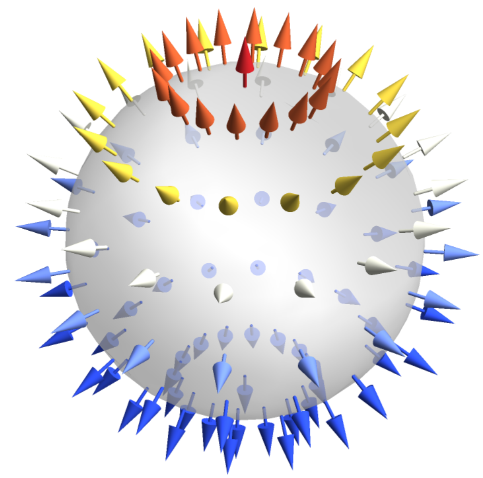
  implications for 2D chiral magnets

# What is a Skyrmion? topological soliton with Quantized topological charge or chirality





"Winding Number" on the unit sphere in spin-space

$$Q = \frac{1}{4\pi}\int d^2\mathbf{r}\ \widehat{\mathbf{m}}\cdot(\partial_x\widehat{\mathbf{m}} \times \partial_y\widehat{\mathbf{m}})$$

$$= 0, \pm 1, \pm 2, \ldots$$

$$\Pi_2(S^2) = \mathbb{Z}$$

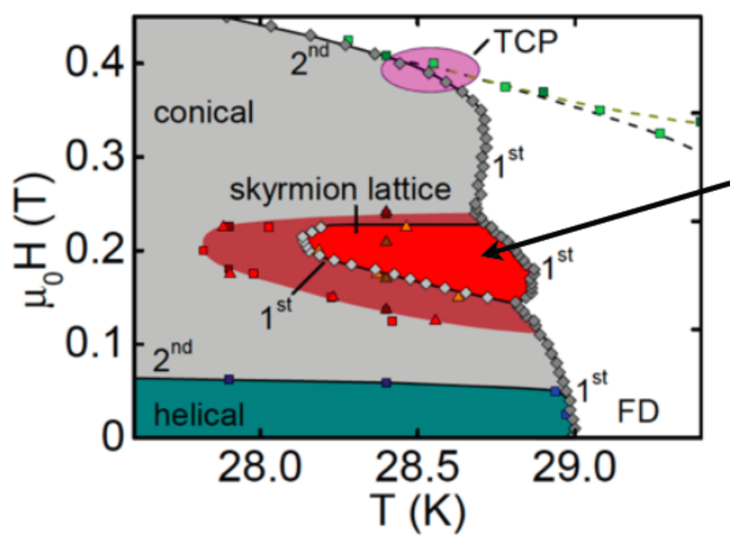
# Magnets with broken inversion
# Exchange + DM → Skyrmion Crystals

Theory: Bogdanov & Hubert, JMMM (1994)
Rossler, Bogdanov, Pfleiderer, Nature (2006)

## MnSi, $Fe_{1-x}Co_xSi$, FeGe, $Cu_2OSeO_3$,…

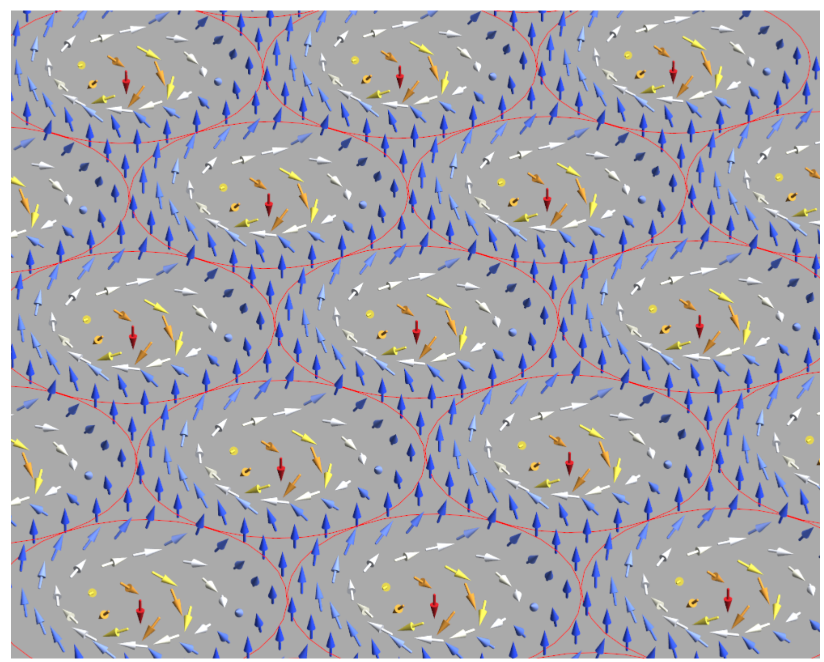
expts: Pfleiderer
Tokura

MnSi $P2_13$




Bauer et al, PRL **110**, 177207 (2013)

## Skyrmion crystal (SkX)



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Theory: Bogdanov & Hubert, JMMM (1994)
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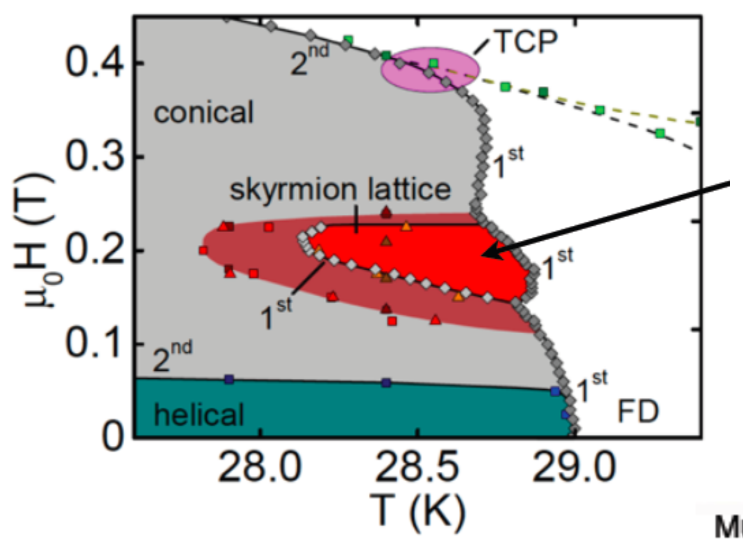
MnSi, $Fe_{1-x}Co_xSi$, FeGe, $Cu_2OSeO_3$,... expts: Pfleiderer
Tokura

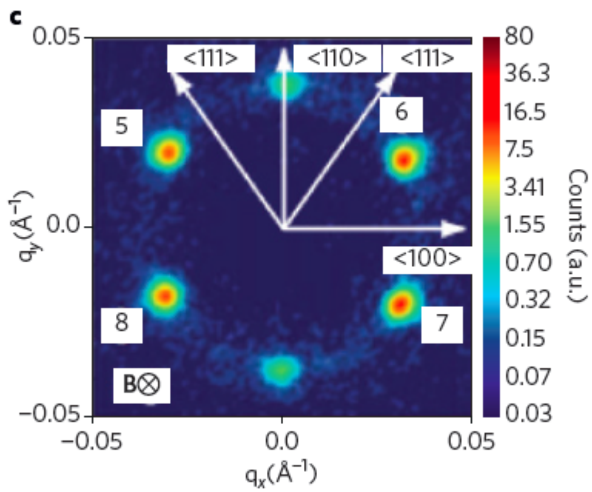
## Skyrmion crystal (SkX) in **q**-space and in **r**-space

MnSi $P2_13$




Bauer et al, PRL **110**, 177207 (2013)

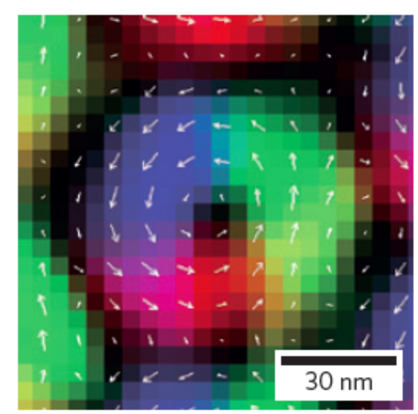
MnSi – SANS




Mühlbauer et al, Science **323**, 915 (2009)

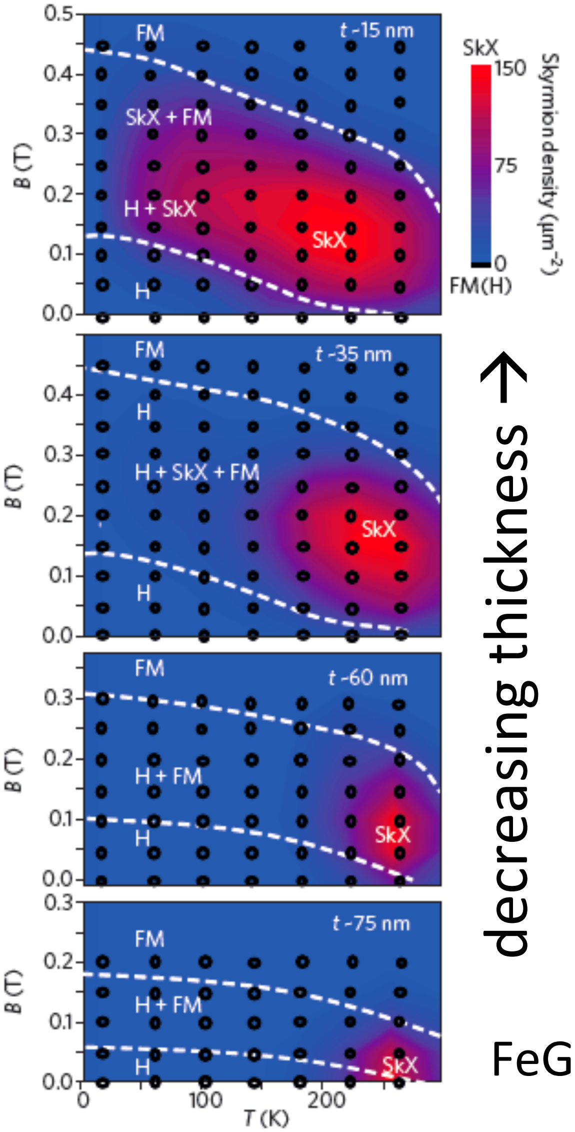
$\lambda \sim 200\mathring{A}$

MnSi – Lorentz TEM




X. Z. Yu et al.,
Nature 465, 901 (2010)

## Enhanced stability in thinner films




FeGe

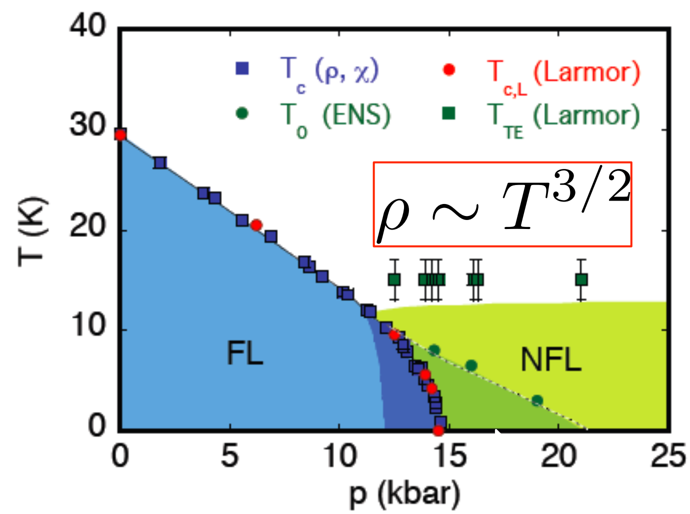
Z. Yu, et. al., Nature Matl 10, 106 (2011)

## Properties of Skyrmion phases

Review: Nagaosa & Tokura, Nature Nano **8**, 899 (2013)

→ emergent "electromagnetism"
→ topological Hall effect
→ low pinning compared to domain walls

→ non-Fermi liquid <u>phase</u>

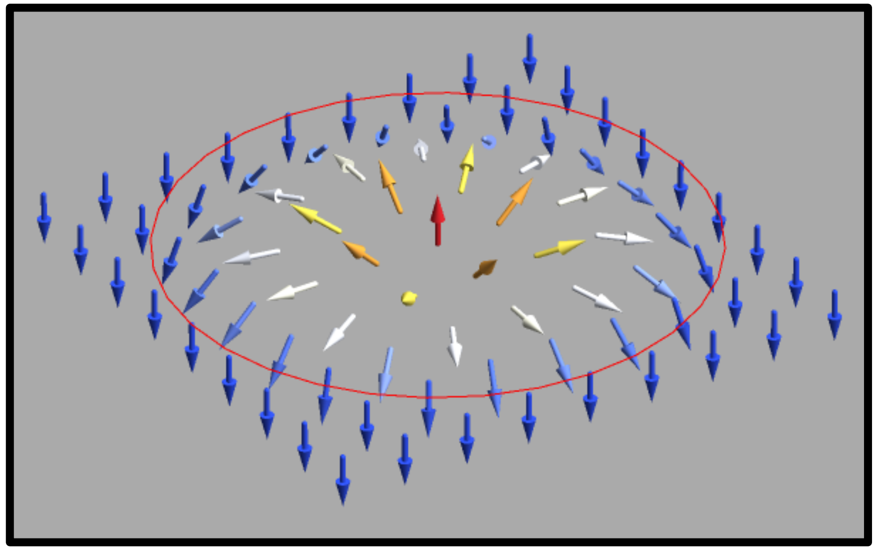



Ritz et al, Nature 497, 231 (2013)

Our focus - motivated by oxide interfaces -

Skyrmions in 2D Chiral Magnets

* Broken inversion
* Rashba SOC
* Role of anisotropy
* $\mathbf{H}||\hat{\mathbf{z}}$

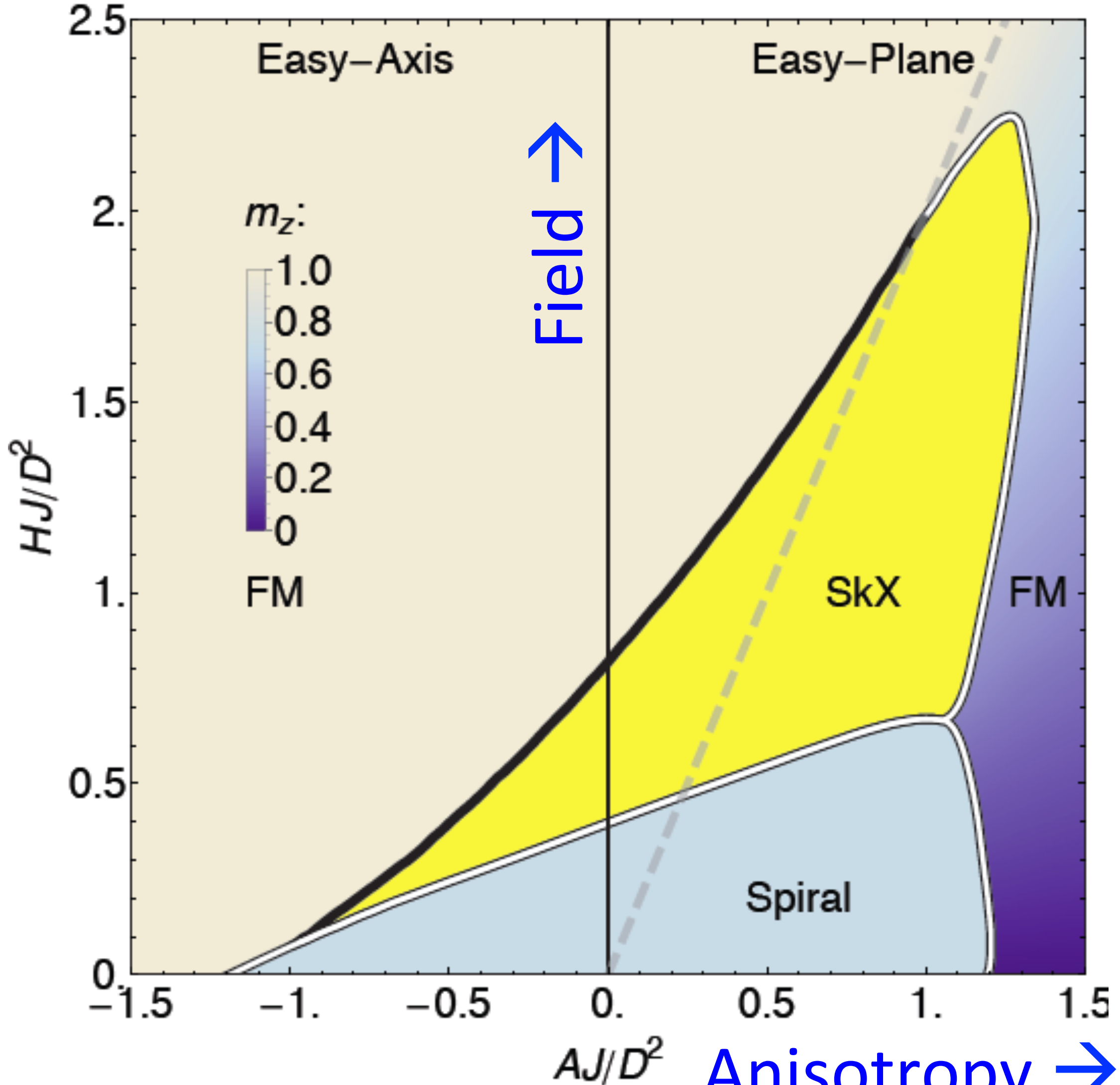


G-L Theory → T=0 Phase Diagram

* Variational calculations
  -- analytical & numerical
* Numerical minimization
  -- conjugate gradient

# T=0 Phase Diagram in (A,H) plane in 2D

* All phase boundaries are 1st order
* Spiral ground state for H=0 and $AJ/D^2 \lesssim 1$
* FM state for sufficiently large H and/or A
* SkX = Skyrmion Crystal




$$A = A_c + A_s$$

Variational calculations (analytical & numerical) <u>and</u> conjugate-gradient minimization

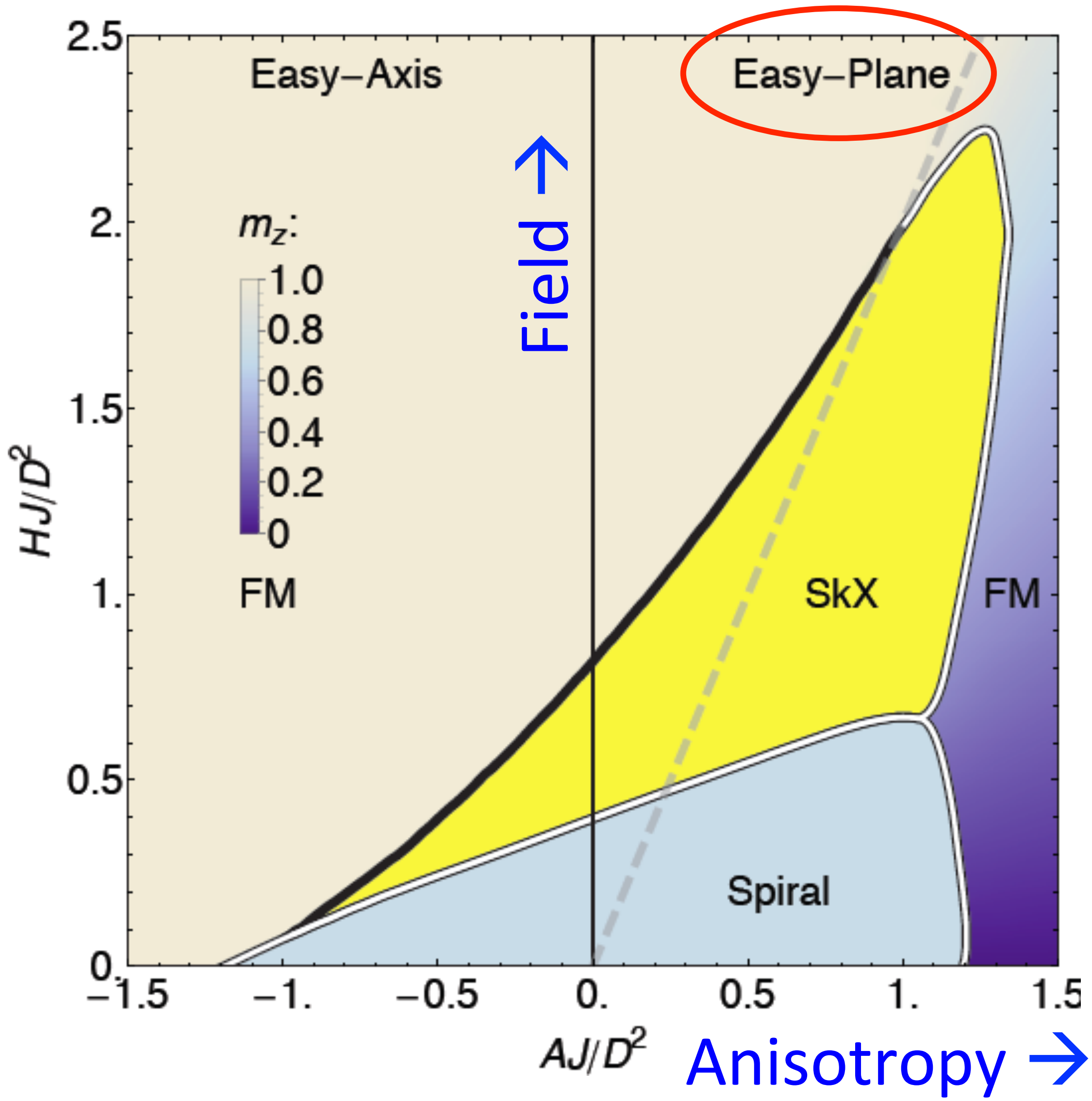
# T=0 Phase Diagram in (A,H) plane in 2D

Surprising stability of skyrmions for easy-plane anisotropy

* natural with compass term

* Very different from 3D for A>0

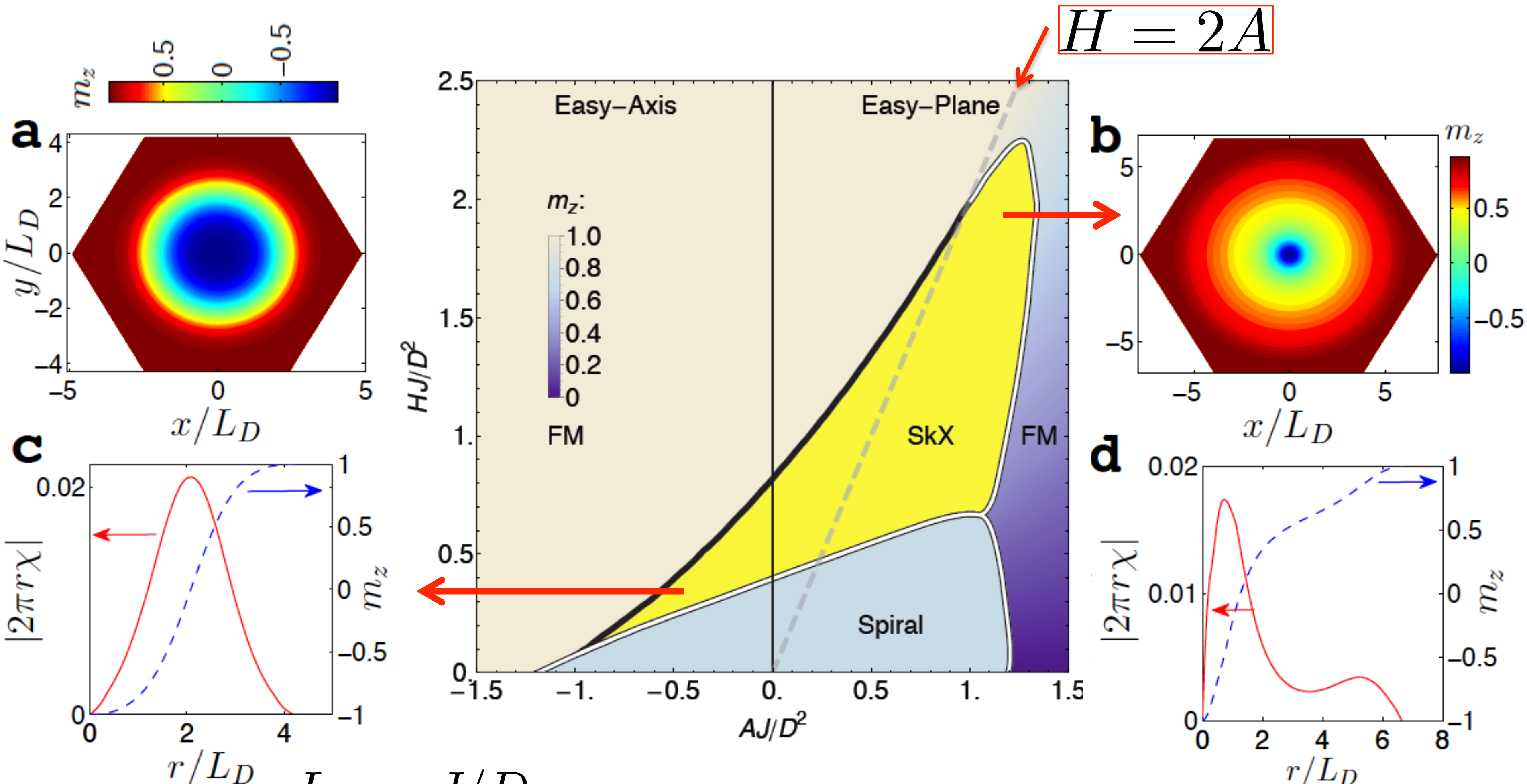
* In 3D SkX only for A<0 (easy-axis)




$$A = A_c + A_s$$

# Why does easy-plane anisotropy favor skyrmions?

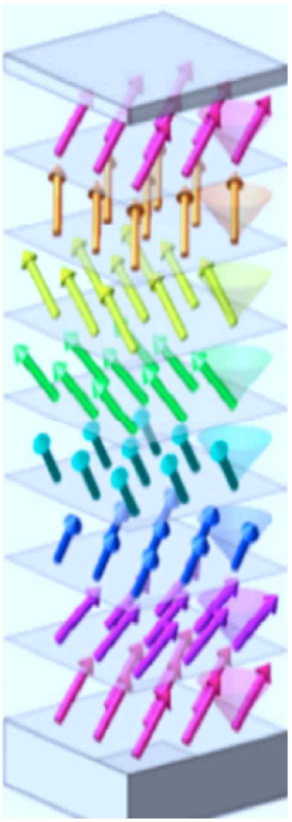
* Compromise between z-field & in-plane anisotropy
* Nontrivial structure of topological charge density

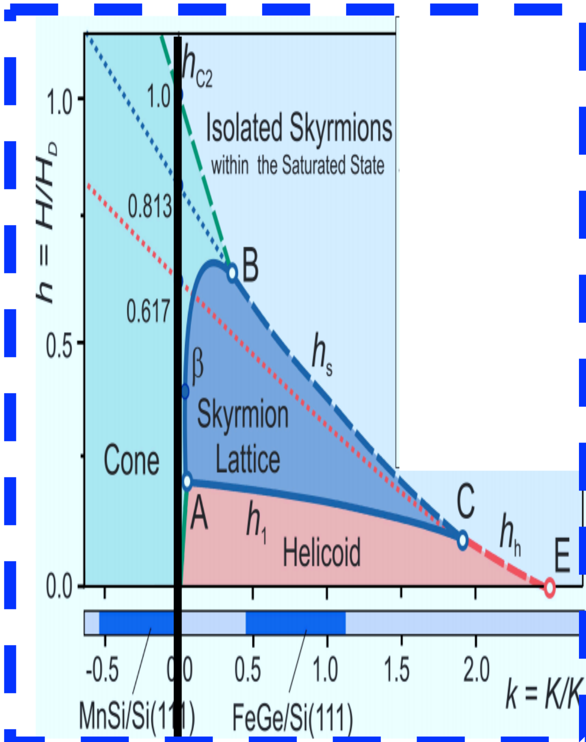


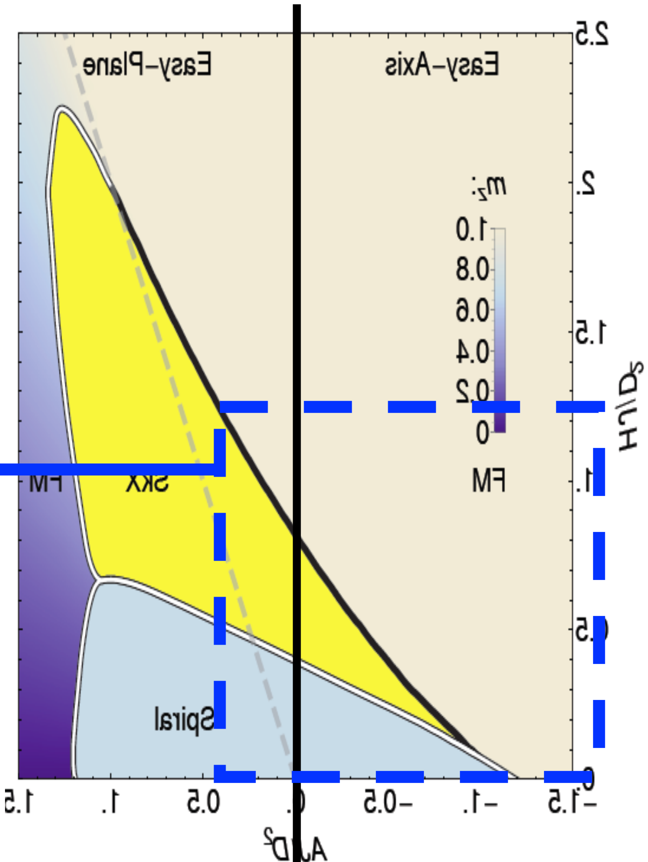

$L_D = J/D$

# 3D vs. 2D

cone





← Easy plane 0 Easy Axis →

← Easy plane 0 Easy Axis →

3D Theory: M. Wislon et al, arXiv:1311.1191

"Cone" phase does not exist in 2D or in quasi-2D Rashba systems

Tokura group → stabilize SkX in 10 nm MnSi films for thickness < J/D

Y. Li et al, PRL 110, 117202 (2013)

# Summary:

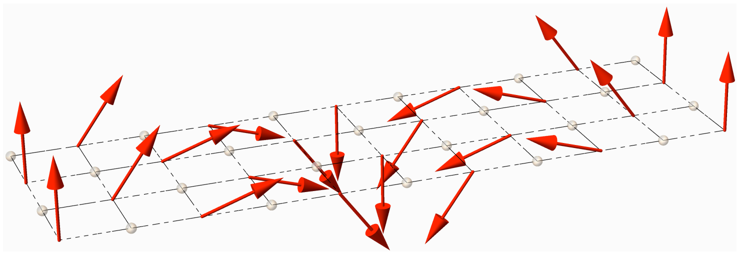
- ❑ Oxide interfaces: evidence for magnetism LAO/STO

- ❑ Broken inversion + Rashba SOC
  Symmetry → chiral DM + compass anisotropy

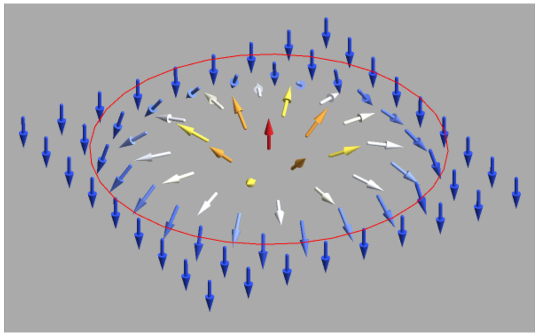
- ❑ Microscopics → $AJ/D^2 = 1/2$

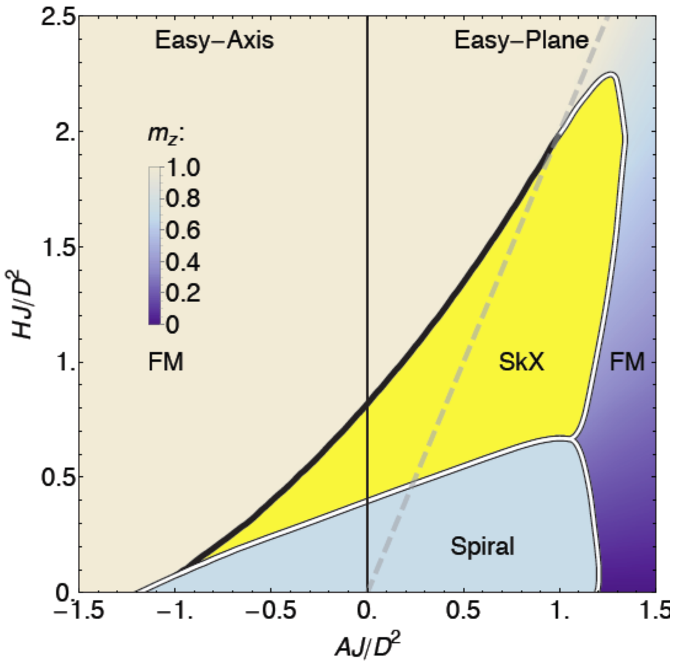
- ❑ Spiral ground state

- ❑ Enhanced stability of
Skyrmion crystal in 2D chiral magnets



Nature Phys. **9**, 626 (2013); arXiv: 1402.7082





The end